

**THE DOE AUTOMATED RADIOXENON SAMPLER-ANALYZER (ARSA)
BETA-GAMMA COINCIDENCE SPECTROMETER DATA ANALYZER**

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ABSTRACT

The Automated Radioxenon Sampler/Analyzer (ARSA) developed at the Pacific Northwest National Laboratory for the Comprehensive Nuclear-Test-Ban Treaty (CTBT) measures four radioxenon isotopes, ^{131m}Xe , ^{133m}Xe , ^{133g}Xe , and ^{135g}Xe . The system produces three sample histograms and three background histograms daily. The analysis of the sample histograms in conjunction with the background histograms is accomplished with a data analyzer that displays various one- and two-dimensional beta-gamma histograms. It also calculates the radioxenon concentrations and the minimum-detectable-concentrations (MDC) for each sample. This paper will describe the software program along with data formats, various software controllable parameters, and calculations.

Key Words: xenon, beta-gamma, software, analysis, CTBT

OBJECTIVE

The remote detection of clandestine nuclear explosions via a nuclear signature is possible due to the large amount of radioactive xenon isotopes generated from fission processes. Because of the very low chemical reactivity of xenon, it can escape the underground blast site through fissures in the ground and be detected hundreds and even thousands of miles from the test site. This feature is the basis behind the radionuclide monitoring portion of the International Monitoring System (IMS) mandated by the Comprehensive Nuclear-Test-Ban Treaty Organization (PrepCom 2000). The IMS has been tasked to establish several global networks for measurement of seismic, hydroacoustic, infrasound, radioactive particulates, and radioxenon concentrations.

To fulfill the CTBT requirement, Pacific Northwest National Lab (PNNL) developed an Automated Radioxenon Sampler-Analyzer (ARSA) to detect the four radioxenon fission products of interest (^{131m}Xe , ^{133g}Xe , ^{133m}Xe and ^{135g}Xe). As the name suggests, the ARSA is an automated system that can operate unattended in remote locations. The system separates and concentrates the ambient and radioxenon from the air and uses a beta-gamma coincidence counting system to determine the activity levels of each of the four isotopes. The collection xenon gas and beta-gamma coincidence counting systems are characterized in detail in several references and hence will not be described in this paper (Reeder et al 1998, Reeder and Bowyer 1998, Bowyer et al 1999a, Hayes et al, 1999).

The ARSA has been deployed in four locations to date; Richland, Washington; the Environmental Measurements Laboratory, located in New York City, during the fall of 1997; DME in Florida during September 1999; and the Institute for Atmospheric Research in Freiburg, Germany, during the fall of 1999 and winter and spring of 2000 (Bowyer et al, 1999b, McIntyre et al 2000). Its most recent performance in Freiburg has been very good and a large data set of radioxenon samples have been collected and analyzed.

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Nuclide	^{131m} Xe	^{133m} Xe	^{133g} Xe	^{135g} Xe
<u>Half-life</u>	11.93 d	2.19 d	5.25 d	9.14 h
<u>Gamma-rays</u>				
Energy (keV)	163.9	233.2	81.0	249.8
Abundance (%)	1.96	10.3	37.0	90.0
<u>X-rays (K-shell)</u>				
Energy (keV)	30.	30.	31.	31.
Abundance (%)	54.1	56.3	48.9	5.2
<u>Beta Spectrum</u>				
Max. Energy (keV)			346.	905.
Abundance (%)			99.	97.
<u>CE (K-shell)</u>				
Energy (keV)	129.	199.	45.	214.
Abundance (%)	60.7	63.1	54.1	5.7

Table 1. Half-lives, principal radiations and abundances of Xe fission products.

Xenon Beta and Gamma Spectroscopy

Before discussing the ARSA Data Analyzer it is first necessary to discuss the salient features of the beta-gamma coincidence spectrum obtained from each of the radioxenon isotopes. Table 1 lists the half-life, prominent gamma-rays, beta endpoint energy, and important conversion electrons (CE) for ^{131m}Xe, ^{133g}Xe, ^{133m}Xe and ^{135g}Xe (Browne and Firestone 1986, National Nuclear Data Center). The two meta-stable xenon isotopes, ^{131m}Xe and ^{133m}Xe, appear in the two-dimensional beta-gamma energy phase space on top of the 30-keV x-ray distribution of ^{133g}Xe. Without beta energy resolution, it would be difficult to unfold these two isotopes from their gamma rays that are not in coincidence with a beta particle or conversion electron (CE). The x-ray and gamma ray of ^{133g}Xe provide two measurements of this isotope, the only one that is specifically called out by the CTBT to have a minimum detectable concentration (MDC, see below) of less than 1.0 mBq/SCM.

Two-Dimensional β - γ Spectrum

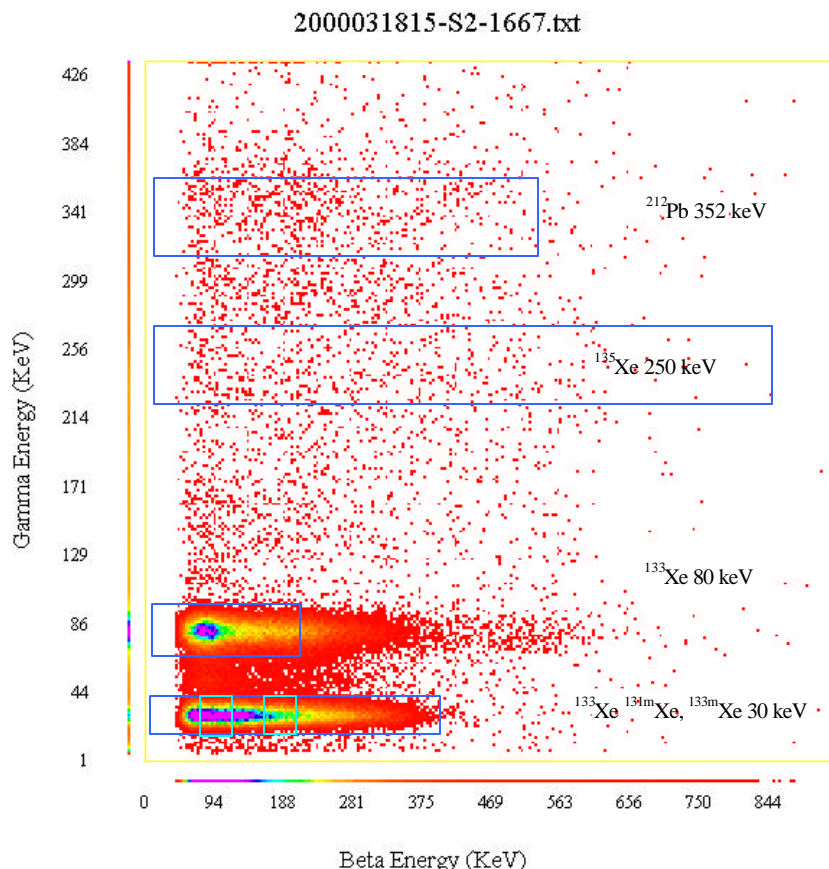


Figure 1. A two-dimensional β - γ coincidence histogram of an intense ^{133}gXe sample taken at IAR. The outlined regions designate 5 radioxenon regions and one region for ^{214}Pb .

The data acquisition computer bins the pulse height data from the beta and gamma Analog-to-Digital-Converters (ADC) in a two-dimensional histogram (see Figure 1). This histogram clearly shows the presence of ^{133}gXe via its 30-keV x-rays and 81-keV gamma-ray. Also indicated are the beta-gamma boxes that outline the regions of interest (ROI's) for each of the four radioisotopes and one region for ^{214}Pb (a short-lived daughter of ^{222}Rn). It is from this histogram and a previous 8-hour background histogram that calculations for the concentration of the four radioxenon isotopes are made as well as calculations for the various MDC values.

The 30-keV gamma ROI has the possibility for $^{131\text{m}}\text{Xe}$, $^{133\text{m}}\text{Xe}$ and $^{133\text{g}}\text{Xe}$ to be present. By carefully gain matching the two beta cell PMT's it is possible to resolve the two conversion electron (CE) peaks of $^{131\text{m}}\text{Xe}$ (129-keV) and $^{133\text{m}}\text{Xe}$ (199-keV) from $^{133\text{g}}\text{Xe}$ (beta plus 45-keV CE). Though it is not present in Figure 1, the radon daughter, ^{214}Pb , interferes strongly with the 81- and 250-keV ROI's (^{214}Pb has x-rays at 79 keV and gamma rays at 242, 295 and 352 keV in coincident with beta particles). The ratio of counts in the 352-keV ROI to those in the 81- and 250-keV regions is constant and therefore it is possible to subtract out the ^{214}Pb contribution in the concentration calculations. The ratios are calibrated by injecting a ^{222}Rn gas sample into the beta cell and measuring the observed ratios.

ARSA Beta/Gamma Analyzer

The ARSA Beta/Gamma Analyzer was designed as a visual basic software tool to evaluate beta-gamma coincidence data produced by the ARSA system. The analyzer is designed to be as flexible as possible, thereby allowing the operator a great deal of freedom in determining how the information is to be presented and utilized. It is based on data files adhering to the RMS 3.0 data format for Beta Gamma systems (Biegalski 2000), and is capable of viewing any file written in this format. However, some capabilities of the analyzer, such as MDC and concentration calculations, have been tailored toward the ARSA system. The analyzer allows two files, a primary (sample) and secondary (background), to be opened at any one time, although the user can elect to open and view any file type described in the RMS 3.0 format (SAMPLEPHD, GASBKPHD, QCPHD, etc.).

The primary form for the analyzer is the Histogram form (see figure 2). This form shows a graphical representation of the detector data and is comprised of 3 main areas:

1. A (upper left panel in Figure 2) two-dimensional, false-color representation of the data with beta energy and gamma energy axes displayed. This image's colors can be scaled linearly or logarithmically to accentuate the structure of the image. Experienced users can immediately determine from this image if further investigation of the data is required. Additionally, potential interference from sources such as Radon will be evident in this image. Both the primary and secondary data files can be displayed.
2. Two one-dimensional histograms that display the Gamma (lower right panel in Figure 2) and the Beta (upper right panel in Figure 2) spectrum on two separate graph controls. A wide range of customization options is available including axis scaling, plotting method, line types, and more. Each graph has zoom-in and zoom-out capability. The graphs can be maximized to aid in viewing the data. Each graph supports multiple traces

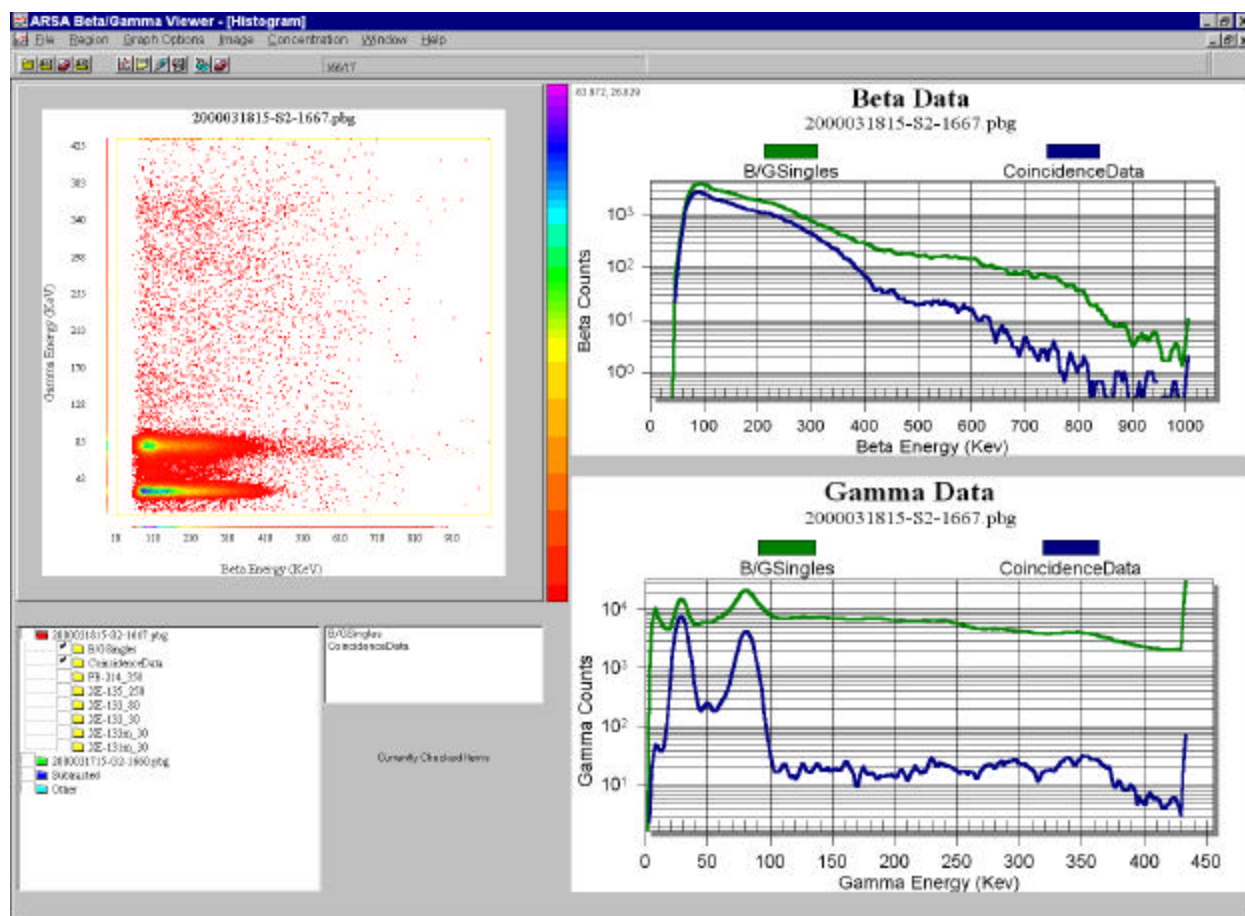


Figure 2. Screen image of the ARSA data Analyzer. See the text for a description.

and allows primary, secondary and subtracted data to be displayed simultaneously. Additionally, the user can select any combination of regions to be graphed. Typically, the entire coincidence spectrum or the Beta/Gamma singles data are displayed.

3. A "region definition area" (lower right panel in Figure 2) displays the filenames that are currently open as well as the regions described in the files. This area of the form is used to determine those regions that are to be graphed. Supplementary regions added by the user are noted in this area and can be graphed as well. Additional menu options allow the user to: switch between primary, secondary, and subtracted views; view /hide the regions of interest; add new regions; and change between grayscale and color.

Additional forms are the header form and the data grid form. The RMS 3.0 data format structures pertinent detector data in blocks and the header form essentially reconstructs these blocks on a tab control so the operator can immediately access pertinent information in the file. If desired, however, the user can elect to view the entire file in raw RMS 3.0 format. Occasionally, the data analysis personnel need to modify specific information contained in the file. These changes are then reflected in this file. The data grid form displays the primary file two-dimensional data in a grid. Additional options have been included to allow the user to add a new region from this form. This form is most useful when the operator is interested in viewing individual counts or is interested in additional information.

Radioxenon Concentration Calculations

The two-dimensional beta-gamma spectrum for the sample and the previous eight-hour background spectrum are used to determine the concentrations of all four radioxenons. The equation takes into account the half-life decay from the beginning of the sampling period, gamma and beta detection efficiencies, possible interferences from ²²²Rn daughters and other xenon isotopes, the appropriate gamma and beta branching ratios, and the xenon gas collection efficiency. The background spectrum provides information on the ubiquitous background as well as the residual xenon and radon gases that adhere to the walls of the plastic scintillator. The background spectrum is subtracted from the sample spectrum for each β - γ bin, accounting for the difference in spectrum collection times (8-hours versus 24-hours). Each ROI is then summed over the beta-gamma energy range that it encompasses to give the total number of counts for that ROI. The radon interference in the 81- and 250-keV ROI's is removed by multiplying the number of counts in the 352-keV ROI by the previously determined radon ratios.

Below is a sample calculation for the ¹³⁵Xe ROI and typical values used:

$$^{135}\text{Xe}_{\text{Conc}} \left(\frac{\text{mBq}}{\text{m}^3 \text{air}} \right) = \frac{C_2}{e_g e_b g_{BR} b_{BR}} \frac{I^2}{(1 - \exp(-IT_C)) \exp(-IT_P) (1 - \exp(-IT_A))} \frac{T_C}{V_{\text{Air}}} 1000$$

where:

C_2	=	Counts -background and radon contamination	→	100.0
e_g	=	g Efficiency	→	49.4%
e_b	=	b Efficiency	→	80.6%
g_{BR}	=	g BranchingRatio	→	90.0%
b_{BR}	=	b BranchingRatio	→	100.0%
I	=	$\ln(2) / ^{135}\text{Xe Half-life (sec}^{-1})$	→	$2.11 * 10^{-5}$
T_C	=	CollectionTime (sec)	→	28800
T_P	=	ProcessingTime (sec)	→	19640
T_A	=	Acquisition Time (sec)	→	86400
V_{Air}	=	cc of Xenon/0.087 cc of Xenon per SCA	→	~ 20.0

Using the above numbers and equation would yield $0.7 \pm 0.07 \left(\frac{\text{mBq } ^{135}\text{Xe}}{\text{SCM}} \right)$ for ¹³⁵Xe and assumes no background counts or ²²²Rn contamination. The factor of 1000 converts from Becquerels to milli-Becquerels.

Calculation of Minimum Detectable Concentration

The minimum detectable concentration (MDC) is a measure of the sensitivity of a particular detector system, and, for the ARSA system, is defined as the lowest amount of activity that can be detected using the counting system given the daily fluctuations in Radon gas interference, the background counts, the memory effect of previous samples on subsequent samples and the detector operations. A good general explanation for the determination of minimum detection limits can be found in reference (Curie 1968), which explains a variety of different measurement scenarios.

For the ARSA system the minimum detectable concentrations calculated assume that a 5% confidence level for measuring a false positive (radioxenon isotopes detected when none are present in the sample) and a 5% confidence level for measuring false negatives (reporting that no radioxenon isotopes are present in the sample when they are) (McIntyre et al 1999). Using these limits, it then is possible to determine the variance of the various backgrounds and interference terms in the absence of the radioxenon isotope and use them to calculate an MDC for the radioxenon isotope in question. Since the previous background count is subtracted from each sample spectrum, its variance must be included. The relevant MDC equation for any of the radioxenons:

$$MDC\left(\frac{\text{mBq}}{\text{SCA}}\right) = \frac{2.71 + 3.21s_0}{e_g e_b g_{BR} b_{BR}} \frac{I^2}{(1 - \exp(-IT_C)) \exp(-IT_p)(1 - \exp(-IT_A))} \frac{T_C * 1000}{V_{Air}}$$

where $s_0 = \sqrt{2 * \text{Background Counts} * (s_{Interference})^2}$, and all other terms were defined in the concentration calculation equation. MDC values for ^{133}gXe from field tests in Germany and New York were typically less than 0.15 mBq/SCA which is well below the 1.0 mBq/SCA requirement for radioxenon monitoring systems. See PrepCom (2000) for a thorough treatment of the MDCs calculated for the ARSA system.

Summary

A software package has been developed to analyze the two-dimensional spectra produced by the ARSA system. This software has proven to be very useful for analyzing data from ARSA systems (and other beta-gamma systems conforming to the RMS 3.0 format) during routine operation and allows operators to calculate concentrations for all of the radioxenons of interest to the CTBT ($^{131\text{m}}\text{Xe}$, ^{133}gXe , $^{133\text{m}}\text{Xe}$ and ^{135}gXe), as well as determining MDCs, and displaying data in raw and graphical forms. The software is flexible and allows significant operator intervention to modify regions of interest, energy calibrations, etc., if desired. The development of this software package was sponsored by the U. S. Department of Energy, Office of Nonproliferation Research and Engineering, Office of Research and Development.

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